Inelastic Quantum Transport

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Abstract

We solve a Schrödinger equation for inelastic quantum transport that retains full quantum coherence, in contrast to previous rate or Boltzmann equation approaches. The model Hamiltonian is the zero temperature 1d Holstein model for an electron coupled to optical phonons (polaron), in a strong electric field. The Hilbert space grows exponentially with electron position, forming a non-standard Bethe lattice. We calculate nonperturbatively the transport current, electron-phonon correlations, and quantum diffusion. This system is a toy model for the constantly branching "wavefunction of the universe".

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This paper is a fundamental study of quantum transport in the presence of inelastic degrees of freedom. We retain coherent quantum effects in a way that has not to our knowledge been done previously. There are a number of relevant model systems, but for concreteness we consider an electron interacting with optical phonons (the Holstein model), driven by a strong electric field. Our calculation is variational. It is non-perturbative in the electron-phonon coupling, the electric field, and everything else.

Most previous work on this problem has relied on rate or Boltzmann equations, which are valid only for weak electron-phonon coupling and ignore quantum coherence effects [1–4]. These theories calculate transition rate probabilities, rather than complex quantum amplitudes. It is not clear how accurate such a treatment is, since, for example, a polaron is a quantum coherent object. This work takes a different approach. We calculate the wave-function $|\psi\rangle$ that is the scattering solution to the Schrödinger equation $H|\psi\rangle = E|\psi\rangle$. The solution consists of the complex amplitude and phase for each basis state in the many-body Hilbert space. The results are relevant to high field transport experiments on semiconductors such as ZnS [5], tunnel cathode structures [6,7], and microchannels [8]. They may also be relevant to systems exhibiting polaron hopping conductivity, such as colossal magnetoresistance (CMR) materials [9]. There has been related theoretical work that takes a very different (path integral) approach to the Fröhlich polaron problem, which has long-range electron-phonon interactions and parabolic bands [10,11].

We consider the Holstein model describing a single electron in a one-dimensional tight-binding lattice, which interacts locally with dispersionless optical phonons [1]. (Other models of inelastic quantum transport can be treated by similar methods.) A constant electric field drives the electron. The Hamiltonian is

$$\begin{aligned}
H = \sum_j \epsilon_j c_j^\dagger c_j - t \sum_j (c_j^\dagger c_{j+1} + h.c.) \\
+ \Omega \sum_j a_j^\dagger a_j - \lambda \sum_j c_j^\dagger c_j (a_j^\dagger + a_j),
\end{aligned} \tag{1}$$

where t is the electron hopping, Ω is the phonon frequency, and λ is the electron-phonon coupling. We consider only the case of zero temperature in this paper. Since there is no

linear response for this problem at $T = 0$, the electric field $\mathcal{E}$ appears explicitly in the Hamiltonian rather than being added as a perturbation. The electric field causes the site energy ϵ_j to decrease linearly with position, $\epsilon_j = -e\mathcal{E}aj \equiv -\alpha j$, where a is the lattice constant and j is the site index. In contrast to linear response theory, the explicit electric field in H allows one to consider the *nonlinear* current carried in a large field. The lattice extends to infinity in both directions, and is translation invariant except that the diagonal site energy decreases linearly with distance.

An electron injected at the origin initially accelerates to the right in the electric field. If the electron-phonon coupling λ were zero, the kinetic energy of the electron would increase until it reached the top of the local band of energetically allowed states. The electron would then reverse direction and undergo Bloch oscillations, with AC but no DC current, characteristic of Wannier-Stark levels [3]. For nonzero electron-phonon coupling, the Wannier-Stark levels decay and the electron continues to move to the right on average, leaving a trail of phonons behind it, like a particle emitting Čerenkov radiation.

We seek to solve as completely as possible the Schrödinger equation $H|\psi\rangle = E|\psi\rangle$, with the Hamiltonian in Eq. (1). This is a formidable problem, because by the time the electron has traveled downhill j sites, it has emitted on the average $\alpha j/\Omega$ phonons. These phonons can be distributed in exponentially many ways on the sites uphill from the electron. Thus the relevant part of the Hilbert space for the electron on site j increases exponentially with j. This problem can nevertheless be numerically solved exactly in an (exponentially growing) variational Hilbert space [12].

We define a variational Hilbert space, which can be systematically enlarged until the solution has converged. A complete set of basis states of the many-body Hilbert space can be written $|M\rangle = |j; \ldots, m_{-1}, m_0, m_1, \ldots\rangle$, where j is the electron position and m_n is the number of phonon quanta on site n. The diagonal energy of the state is $E_M = -\alpha j + \Omega(\sum_n m_n)$. A variational space is defined as follows. A basis state is retained only if: (1) The number of phonon quanta on a given site does not exceed a constant n_{max}. (2) The diagonal energy E_M of a many-body basis state $|M\rangle$ lies in a given energy range,

$\Delta_1 \leq E_M \leq \Delta_2$. (The accuracy is greatest if the desired energy eigenvalue E is near the center of the range.) These constraints are not terribly restrictive, as n_{max} and the energy range can be systematically increased to check that the solution has converged. For technical reasons that will be explained below, we also apply a third condition: (3) all phonon excitations may appear only on the same site or left (uphill) of the electron. This condition is motivated by the fact that for a strong electric field, by analogy with Čerenkov radiation, almost all of the phonons are left behind the electron. This condition can also be relaxed, but with some effort.

To illustrate the method, we first consider one of the simplest nontrivial cases, with $\alpha = \Omega = \Delta_2 = -\Delta_1 = 1$, shown in Fig. (1) [13]. The electron is injected into state $|1\rangle$, which has the electron at the origin and no phonon excitations. It can either hop to the right (state $|3\rangle$), or create a phonon (state $|2\rangle$) [14]. The lattice extends to infinity, with the number of possibilities increasing exponentially as the electron moves downhill. The off-diagonal matrix elements are $-t$ on bonds a and b, $-\lambda$ on bonds x and y, and $-\lambda\sqrt{2}$ on bond z. The diagonal energies are -1, 0, or $+1$. To take a specific example, the Schrödinger equation on site (4) is

$$E\psi_4 = \epsilon_4\psi_4 - t\psi_2 - t\psi_8 - \lambda\psi_6, \tag{2}$$

where the diagonal energy $\epsilon_4 = 0$. The exponential growth in the number of states should be contrasted with the standard polaron problem where the electric field $\mathcal{E} = 0$, which is shown in Fig. (2).

We seek a scattering solution for the lattice of Fig. (1) where the electron is injected on site 1, and generalized currents flow along the bonds. The Schrödinger equation is satisfied on all sites (other than 1) with an energy eigenvalue E that is known in advance. The lattice has no loops, and is in fact an unusual type of Bethe lattice. The entire lattice connected downstream (to the right) of any b bond is the same as that for any other b bond. This leads to the condition that in the scattering eigenfunction, the amplitude ratio across any b bond is a fixed complex number that will be called b, $\psi_3/\psi_1 = \psi_8/\psi_4 = \ldots \equiv b$. The same

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can be said of x bonds, etc. [16]

The scattering problem can be solved by taking a finite piece of the lattice, as shown in Fig. (1). Consider a terminal site j, whose downstream neighbor m on a b bond has been omitted. The Schrödinger equation on site j is written assuming $\psi_m/\psi_j = b_0$, where b_0 is an initial guess for b. Proceeding similarly for the other terminal sites, the system is solved for all amplitudes ψ_i. A new set of complex numbers $a_1, b_1, \ldots$ is obtained by taking ratios of the amplitudes on specified interior bonds. This new set of complex numbers is then used on the edge sites, and the equations iterated until self-consistency is achieved. The initial complex numbers are chosen with a positive imaginary part, corresponding to outgoing solutions. The solution is obtained numerically by solving a sparse complex system of equations of the form $Ax = b$. (Since the eigenvalue E is known in advance, the numerical routine required is for simultaneous linear equations [17], not for eigensystems.) Once the wavefunction has been obtained [18], the expectation of any observable can be calculated. If the maximum number of phonons per site in condition (1) or the energy range in condition (2) are increased, the representation of the problem still has no loops. The graph does, however, contain more inequivalent bonds. We have solved this problem with hundreds of inequivalent bonds (rather than the 5 types discussed above) by the same iterative method [19]. If condition (3) above is relaxed, and phonons are allowed to the right of the electron as well, then a low density of loops appears in the graph. The loops lead to complications that will be discussed below.

It is sometimes asserted in transport theory that an electron scattering from a static impurity retains its phase, but that when a phonon is emitted or absorbed, the phase is randomized. We do not subscribe to this point of view. Every many-body basis state in Fig. (1) has a wavefunction ψ_j with both an amplitude and definite phase. Some observables, such as the electron density on a particular site, make no use of the phase information in this basis—they would be unchanged if all phases were randomized. Other observables, however, depend crucially on the phases.

Consider the average displacement of the oscillator on site j when the electron is on site

m, $\langle(a_j^\dagger + a_j)c_m^\dagger c_m\rangle$, plotted in Fig. (3). This expectation depends crucially on the relative phases of the different ψ_i. It is straightforward to show that as the real physical distance $m - j$ between the electron and phonon in the correlation function increases, the expectation requires knowledge of the relative phases of many-body basis states that are farther apart in the Hilbert space, Fig. (1). (The metric distance l between basis states in the many-body Hilbert space shown in Fig. (1) is defined as the minimum number of bonds to get from one to the other.) Although distant phase correlations are required for large $m - j$, the sum of the many terms is found to be small. In fact, for large $m - j$, one would not obtain a seriously wrong answer by taking the computed amplitudes and averaging over all random phases, which would give zero for the correlation function.

Also plotted in Fig. (3) is the phonon energy on site j when the electron is on site m, $\langle a_j^\dagger a_j c_m^\dagger c_m\rangle$, which goes to a constant for large $m - j$ (except near the injection site). The electron sets a phonon ringing as it passes. As the electron moves far away, the energy remains in the oscillator (it has nowhere to go), but the oscillator is as likely to have a positive as a negative displacement. These phonon excitations should be measurable by neutron scattering. The correlations plotted as triangles in Fig. (3) cannot be calculated by the methods of Ref. [1]. (It has generally been assumed in previous work that the phonons begin and remain in equilibrium.)

We now consider the current or average electron velocity. Figure (4) plots the velocity $v(E) = \langle J(E)\rangle/\langle n(E)\rangle$, the ratio of the particle current to the particle density, as a function of the total energy eigenvalue E. The solid curves are for different values of the electron-phonon coupling λ, and the dashed curve is for a smaller electric field $\alpha = 1/2$. The velocity is a periodic function of the energy E with period α, because a shift in the energy by α is equivalent to a shift in the origin by one lattice constant. The dashed curve has period $1/2$, and is more nearly constant than the others. The fact that the numerically calculated $v(E)$ is fairly accurately periodic is a confirmation that the variational energy range $[\Delta_1, \Delta_2]$ is large enough that the answer has converged for the energy eigenvalues E

plotted. For small electron-phonon coupling λ, there are energies E (the eigenvalue of the entire coupled electron-phonon system) where the electron cannot propagate at all. This is analogous to band gaps in crystals. The forbidden regions are perhaps less surprising when one considers that if the electron-phonon coupling λ were zero, the electron would be localized in a Wannier-Stark state, and there would be no current at any energy. We believe that these forbidden regions would disappear if the optical phonons were given nonzero dispersion.

It is more difficult to calculate the velocity $v(\alpha)$ as a function of the electric field, because this is a nonanalytic function. When α/Ω is a low order rational, the representation of the Hilbert space as shown in Fig. (1) contains subsets that are 1d periodic tight-binding lattices with a small basis, such as the branch $byby\ldots$. In contrast, for α/Ω irrational, all such subsets are quasiperiodic, leading to different transport. A similar argument accounts for the energy dependence of v when α/Ω is rational: it matters how close the diagonal energies are to the eigenvalue E. Some aspects of these results differ from those obtained previously. Using rate equations that neglect quantum coherence, Emin and Hart (and others) obtain an infinite drift velocity for dispersionless optical phonons [1], in contrast to the finite result of Fig. (4). We are, however, in qualitative agreement with Ref. [1] in that reducing the electric field in this regime increases the current. We believe that the enhanced nonlinear conductivity measured by Maekawa in ZnS occurs at rational values of α/Ω. In contrast to his interpretation [5], this need not signal the existence of long-lived Wannier-Stark levels; see also Ref. [3].

We now consider the time evolution of a wavepacket, formed by injecting an electron into site (1) with $\psi_1 = \exp(-iE_0t - t^2/2\tau^2)$. For a packet that is narrow in energy space (wide in real space), the time evolution depends on the first and second energy derivates of the amplitude and phase of the complex numbers $\{a, b, x, \ldots\}$. A detailed calculation [20] shows that the width increases in time as

$$\langle [x(t) - \bar{x}(t)]^2 \rangle = \sigma_0^2 + \mu t + \nu t^2. \tag{3}$$

The last term is a quantum coherent spreading similar to the one that appears in a simple 1d chain (without branches), with ν decreasing as the initial wavepacket gets wider, $\nu \sim \sigma_0^{-2}$ [21]. The additional term is linear in time, with the coefficient μ independent of the initial width of the wavepacket. It describes the same behavior as the *classical* diffusion of particles due to Brownian motion. It is perhaps surprising to see such behavior emerge from a fully coherent quantum mechanical calculation (without disorder) at zero temperature. One reason the μt term appears in the branching Hilbert space is that the energy variation of the modulus of the complex numbers causes parts of the wavepacket with slightly different energies to move preferentially into different branches of the tree. (This property is not present in standard Bethe lattices [22].) Once on different branches, these amplitudes cannot destructively interfere, which causes the wavepacket to spread [23]. Another reason is that the group velocities differ on different branches.

Another question is how much entropy is generated as the electron travels downhill. The technical answer is that no entropy is generated. The system begins in a pure state and remains in a pure state under time evolution of the Schrödinger equation [24].

When phonon excitations are allowed to the right of the electron (relaxing condition 3), the structure of the Hilbert space shown in Fig. (1) acquires extra states that form infrequent loops. The structure is no-longer exactly tree-like, but it is still self-similar. The problem can still be solved by similar methods (finding "images" of the terminal sites nearer the origin of the lattice), and also by novel methods [20]. None of these methods is as yet completely satisfactory, however. One problem is that the iterative scheme may fail to converge for certain energies. We will not discuss this extension in any detail in this publication.

A final point concerns the foundations of quantum mechanics. The "wavefunction of the universe" has been described as residing in an abstract space that is constantly branching [25]. One question that has been considered is the issue of why amplitudes on different branches seem not interfere with each other. The usual answer is known as the process of decoherence [26]. The Holstein polaron in an electric field, with a Hilbert space illustrated in Fig. (1), provides a simple toy model for investigating such questions. As described above,

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there is noticeable interference at least for short times for the Holstein model.

FIGURES

FIG. 1.   A variational many-body Hilbert space for an electron coupled to phonons in an electric field. Each dot represents a basis state in the many-body Hilbert space. Lines represent nonzero off-diagonal matrix elements. The many-body problem can alternatively be viewed as a 1-body problem, where the dots are Wannier orbitals with hopping matrix elements shown by the bonds.

FIG. 2.   A variational many-body Hilbert space analogous to Fig. (1) is shown for the case of zero electric field. Only states with 0 or 1 phonon excitation on sites not farther than one lattice spacing from the electron are included. State $|1\rangle$ has no phonon excitations. Vertical bonds create phonons, and (nearly) horizontal bonds are electron hops. For $\mathcal{E} = 0$, the lattice is translation-invariant, does not grow exponentially, and can be solved simply in $\vec{k}$-space [15] .

FIG. 3.   The oscillator coordinate x_j when the electron is on site m, $\langle (a_j^\dagger + a_j) c_m^\dagger c_m \rangle$, plotted as a function of j for $m = 8$ (triangles). The oscillator energy $\langle a_j^\dagger a_j c_m^\dagger c_m \rangle$ as a function of j (squares). The electron is injected into the system on site 0. Parameters are $\lambda = 0.5$, $\alpha = \Omega = t = 1$, $[\Delta_1, \Delta_2] = [-3, 3]$, $E = 0.6$, with up to three phonons per site.

FIG. 4.   (a) The drift velocity v (current/density) is plotted as a function of the energy eigenvalue E for electron-phonon coupling constant $\lambda = 0.3, 0.5$, and 0.7, from the bottom curve up. For the smaller $\lambda's$, there are forbidden energies at which there are no propagating states. Other parameters are $\alpha = \Omega = t = 1$. Basis states are retained in the diagonal energy range $[\Delta_1, \Delta_2] = [-4, 4]$, with up to two phonons per site. The dashed curve is for electric field $\alpha = 1/2$, $\lambda = 0.5$. It has been shifted up by 0.15 .
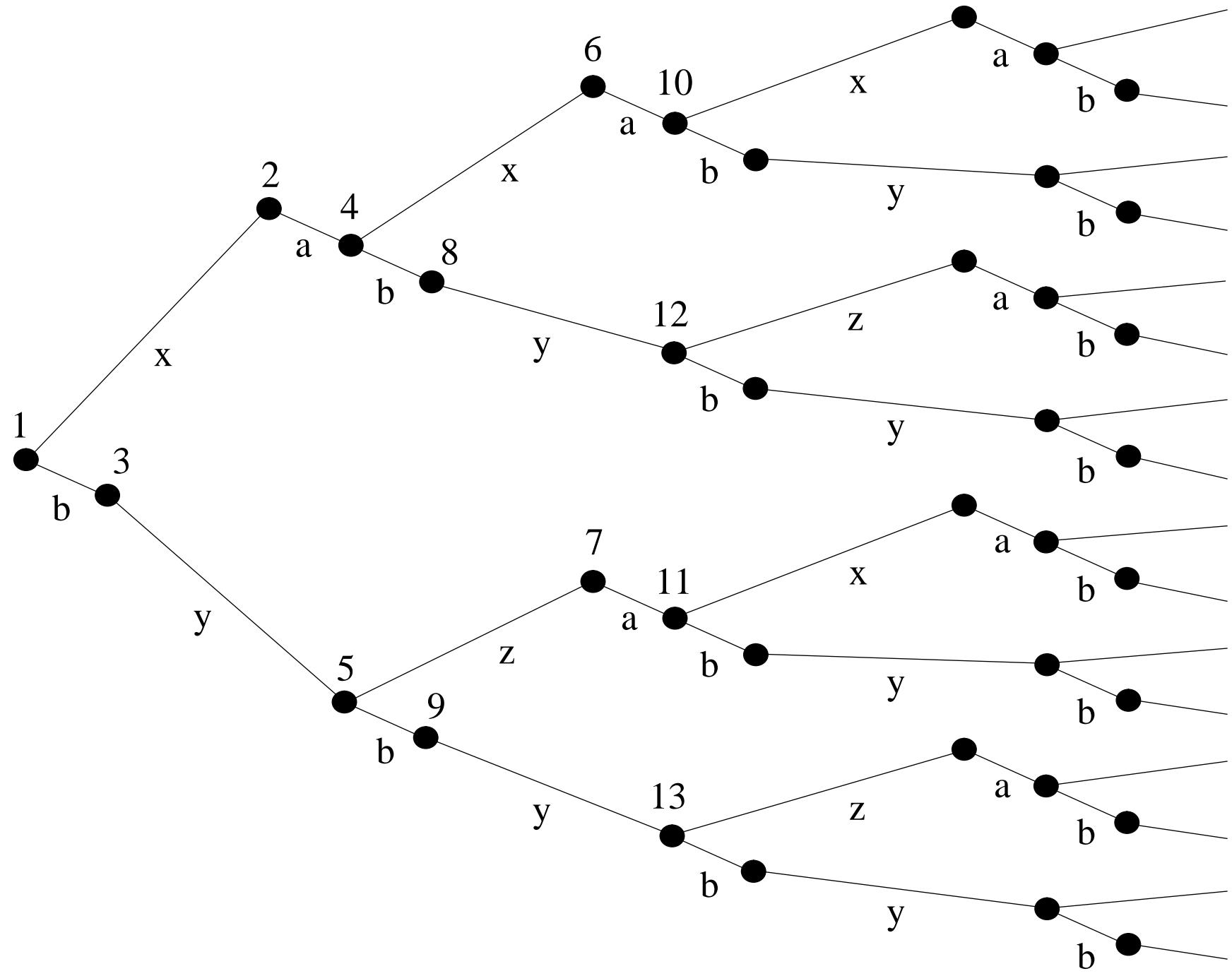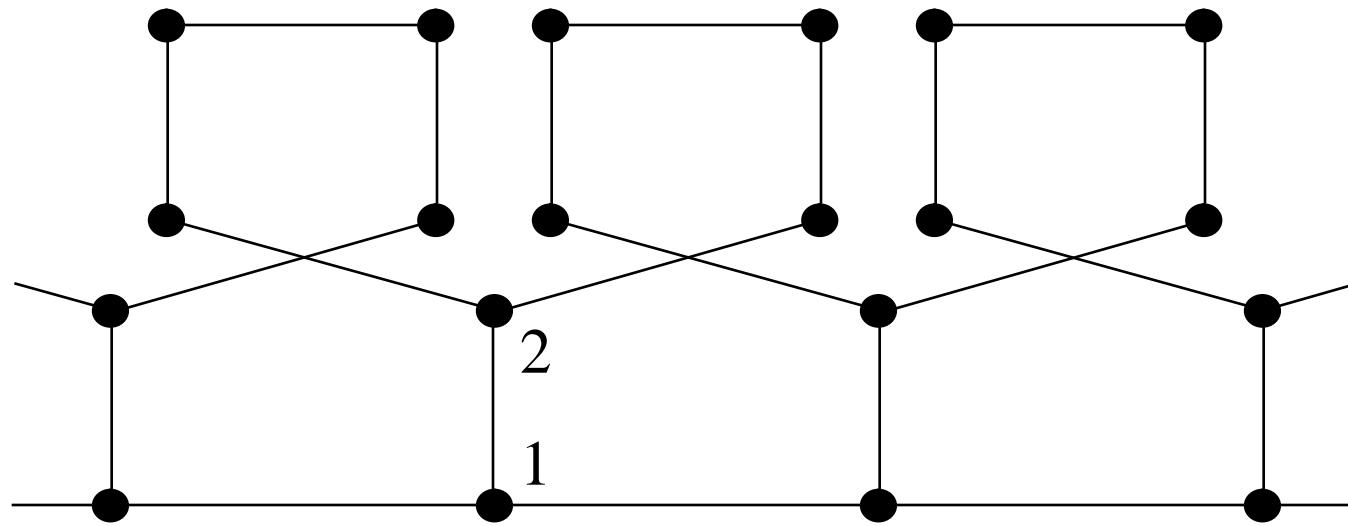
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[12] In a simpler problem relevant to resonant tunneling diodes, phonons are restricted to a small region of space, and exponential growth does not occur. See Janez Bonča and S. A. Trugman, Phys. Rev. Lett. **75**, 2566 (1995).

[13] A field $\alpha = \Omega$ is a very large electric field, corresponding to a potential drop of an optical phonon frequency per lattice site. Experiments are, however, done in this regime [5–7]. One could alternatively consider a site in the Hamiltonian to represent a cluster of several atoms, which would reduce the physical electric field.

[14] The electron could also hop to the left, but this dead-end branch does not affect the results, and is omitted.

[15] S. A. Trugman, in *Applications of Statistical and Field Theory Methods to Condensed Matter,* ed. D. Baeriswyl et al., (Plenum, 1990).

[16] The infinite tree attached downstream from a given site can be regarded as a lumped impedance, similar to an antenna problem.

[17] Z. Zlatev et al., Computer Science, Y12M Solution of Large and Sparse Systems of Linear Algebraic Equations, Documentation of Subroutines, ed. G. Goos et al., (Springer-Verlag, 1981).

[18] If each site in Fig. (1) is labeled by the sequence of bonds that are followed to reach it, the question "what is the wavefunction on site j?" answers itself. For example, $\psi_{11} = \psi_{byza} = byza$, the product of the self-consistent complex numbers.

[19] A unique solution is always found for outgoing boundary conditions. The complex numbers $\{a, b, \ldots\}$ are the same on all equivalent bonds, and do not change to machine accuracy if the lattice is expanded to include additional generations.

[20] Janez Bonča and S. A. Trugman, unpublished.

[21] It is clear from Fig. (4) that since the wavepacket is composed of a range of energies, and each energy has a different velocity, the width-squared $\langle [x(t) - \bar{x}(t)]^2 \rangle$ must increase as t^2 in the long time limit.

[22] E. N. Economou, *Green's Functions in Quantum Physics*, Springer-Verlag, 1990.

[23] In the simple 1d chain, one can write the wavefunction as a sum of plane waves. Each plane wave has infinite spatial extent; the sum has a finite extent because of destructive interference.

[24] We have, however, calculated a basis-dependent "entropy" $\tilde{S}$ as the Shannon entropy in the phonon sequence left behind, assuming that all phase information has been lost. The result is $\tilde{S} = \gamma N$, where γ is a constant that depends on coupling constants.

The calculation is somewhat complicated by the fact that the probability of finding m_j phonons on site j depends on m_{j-1}. We emphasize, however, that $\tilde{S}$ is not a true entropy and that the value of $\tilde{S}$ is basis-dependent.

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phonon site j